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On the Crystal and Magnetic Behavior of $ScFe_4Al_8$ Single Crystal

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Nuclear and magnetic properties of the $ScFe_4Al_8$ single crystal were found to exhibit unparalleled complexity of nuclear and magnetic structures. Our previous neutron measurements revealed presence of two modulation vectors, both along [ε, ε, 0], however with different critical temperatures. Recent experiments forced us to revise our knowledge of the structural ordering in the sample. So far, the crystal structure of this alloy, being of $ThMn_{12}$-type, has never been questioned.

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1. Introduction

The ternary compounds with the general formula MFe_4Al_8 (M = An, RE, Sc, etc.) are known to crystallize in the body centered tetragonal $I4/mmm$ symmetry (space group no. 139). They form large family of intermetallics showing a variety of magnetic structures, and in consequence of physical properties. Our earlier studies of crystal and magnetic structures of (U, Th)$(FeAl)_{12}$ samples showed identity of crystal structures and certain magnetic similarity in both series of actinide's systems. In the case of $ScFe_4Al_8$ compound these rules turned out to fail. Our recent experiment, whose details are presented in this paper, showed that a single crystal of $ScFe_4Al_8$ discloses very surprising effect of two- or three--dimensional nuclear cell doubling. So far, in the analysis of the neutron data, we had no reason to doubt that in this perfectly ordered crystal structure (see Fig. 1) the position 2a is occupied by Sc, the site 8f by Fe, whereas the sites 8i and 8j by the Al atoms [1]. During systematic investigation, the unit cell dimensions of $Sc(FeAl)_{12}$ series have never exceed 8.65 Å × 8.65 Å × 5.04 Å values. It was found earlier [1] that the magnetic structure is described by two modulation vectors, both, however, seemingly along same [110] direction: $k_1 = \{\varepsilon_x, \varepsilon_x, 0\}$ and $k_2 = \{-\varepsilon_x, \varepsilon_x, 0\}$, where $\varepsilon_x = 0.18$ and 0.13. Both modulations exhibit different temperature onsets. The mode 1 with $\varepsilon_x = 0.18$ vanishes above 115 K, and mode 2 with $\varepsilon_x = 0.13$ vanishes at ≈ 220 K. The modulations found strongly suggested presence of double cycloid spiral (DCS) type. The modulation vectors turned out to be weakly dependent on temperature and led to weakly canted antiferromagnetism within single unit cell. In light of presumed magnetic inactivity of scandium, the

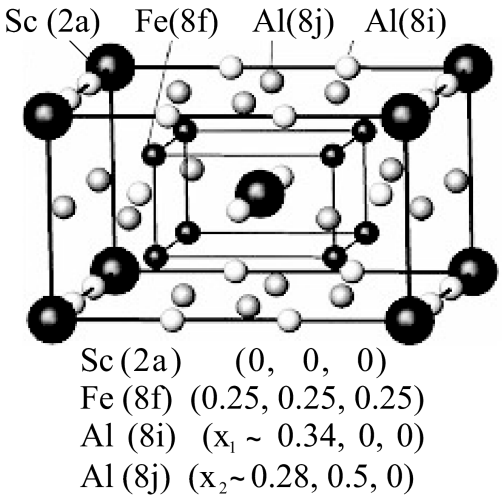

Fig. 1. Crystal structure of $ScFe_4Al_8$ consisting 26 atoms (two molecules) per unit cell $I4/mmm$ with the $a = 8.623(5)$ Å and $c = 5.008(4)$ Å cell parameters at 8(2) K [1].

iron atoms must play key role in formation of the incommensurate magnetic structure [1]. Because the magnetic structure connected with the two modulation vectors was not fully recognized, especially the one connected with the second mode, it was interesting to carry out the experiment which would have resolved the problem. To the authors' best knowledge, such a double modulated structure with the propagation vectors along the same direction has never been observed in MFe_4Al_8 compound.

2. Experimental details

A single crystal of Sc_4Al_8 was obtained in Katowice from stoichiometric amounts of the elements, which were re-melted several times in a Czochralski equipment. The single crystals were subsequently extracted from the levitated melt at a velocity of about 0.1 mm/min. The best crystal needle has a cylindrical shape with a length of about 8 mm and diameter of 2 mm.

The Laue diffraction experiment carried out on this single crystal needle was set in order to check both findings and to complement magnetic structure of the intermetallic alloy by recent study carried out in the temperature range 5–250 K on the Very Intense Vertical Axis Laue Diffractometer (VIVALDI, ILL) using a "white" neutron beam. The diffracted intensities were recorded on a cylindrical image plate [2].

An additional test using hard X-rays facility (100–400 keV) was also carried out at the ILL.

3. Results and discussion

The Laue diffraction measurements carried out at 5 K intended to bring more data on magnetic structure. Thus, on the first steep we made an effort to describe the Laue pattern in expected nuclear structure. As it turned out, only some of the experimentally observed spots fulfill the conditions of the appropriate symmetry. Particularly, the spots localized at the Laue photograph center, which are not indexed within body centered cell ($a = 8.63$ Å and $c = 4.96$ Å) require bigger unit cell dimensions. Noteworthy, all of the extra peaks have the same shape as those indexed, which means that they are coming from the same part of the crystal, which allow to discard a possible polycrystalline origin. A folding of the unit cell, keeping its tetragonal body-centered character, but with a doubling of the three cell parameters ($a = 17.26$ Å and $c = 9.92$ Å) allowed us to completely simulate the experimental diffraction patterns.

Unfortunately, there is also another unit-cell and symmetry, which can describe Laue's data equally well, namely an orthorhombic primitive unit cell with the dimensions: $2a \times a \times 2c$. The neutron experiment carried out on VIVALDI turned out to be inconclusive with regard to the exact determination of the general crystal structure. Thanks to a Laue hard X-ray test, using an energy dispersive technique, two intense Bragg reflections were measured at low diffraction angles (Fig. 2). They correspond to d-spacing of 4.3 and 2.7 Å and could be interpreted as respectively the (200) and (310) reflections of a $I4/mmm$ structure with $a = 8.63$ Å and $c = 4.96$ Å. However, in both cases, two weak but significant additional reflections were observed at low energy. They correspond to longer d-spacing, forbidden with the I symmetry: $a \times a \times c$ unit-cell, but which can be easily explained by a cell doubling along both a and b directions. Because of the equatorial geometry of the diffraction experiment, this technique cannot afford information about the c direction.

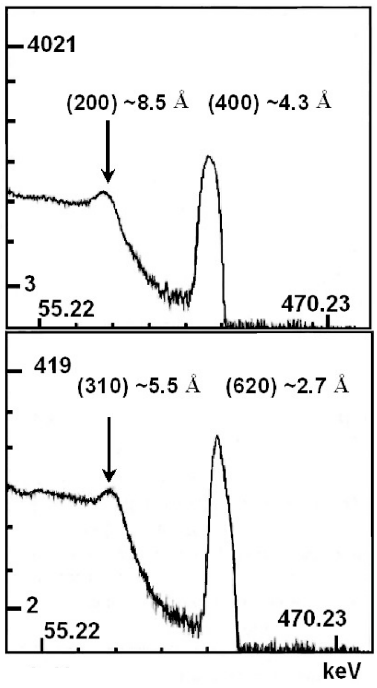

Fig. 2. The intensities (shown in logarithmic scale) vs. energy show two Bragg reflections and their harmonics. Presence of (310) reflection (lower part) indicates that the cell parameters' doubling takes place at least in the basal plane.

Whilst a doubling of an atomic unit-cell axis is very evident in the Laue patterns, this is less so when the magnetic unit cell corresponds to a doubling of an axis. In agreement with previous findings, the well-visible magnetic satellites indicated presence of modulation with wave vectors of the $k = \{\varepsilon_x, \varepsilon_x, 0\}$ type. However, they turned out to appear not only around the allowed nuclear reflection $h + k = 2n$, $l = 2n$ but also around reflections with $l = 2n + 1$ (see Fig. 3).

The additional magnetic reflections were observed along $(h\bar{2}1)$, $(\bar{1}k1)$ and $(h32)$ lines. All observed satellites form typical quartets disappearing above 100 K. This means that all of them have to be connected with the single modulation previously characterized as $k = \{0.18, 0.18, 0\}$. The second mode, namely $k = \{0.13, 0.13, 0\}$ type, perhaps due to the low intensity of peaks, have not been detected. This is quite puzzling

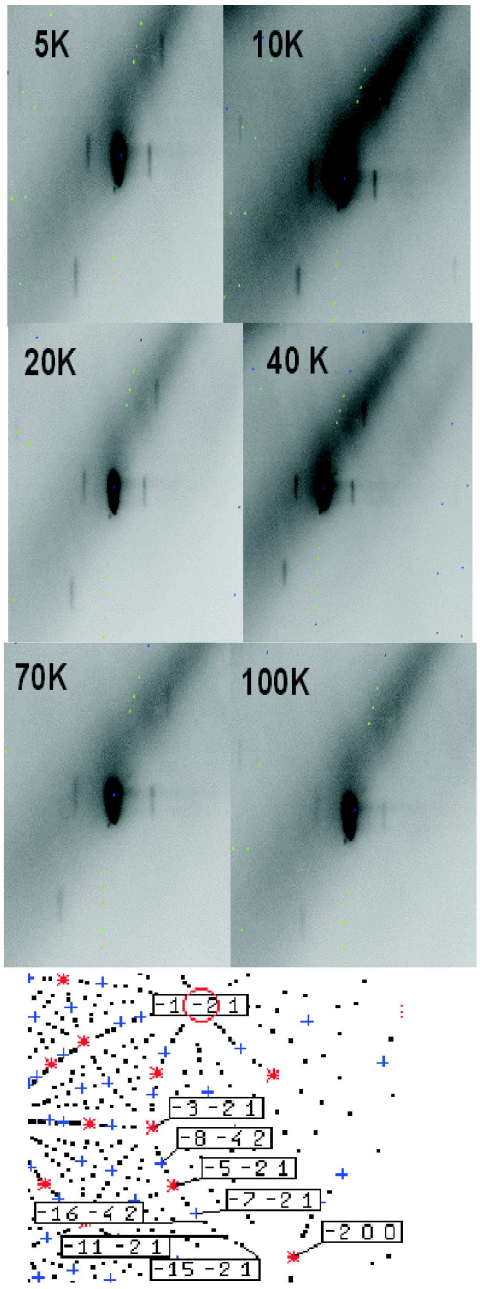

Fig. 3. Main nuclear spot $(\bar{2}00)$ to which four magnetic satellites: $(\pm\varepsilon,\ \pm\varepsilon, 0)$ vanishing above 100 K are attached. Similarly, not-allowed but well-visible magnetic satellites of $(\bar{1}k1)$ nuclear reflections were observed.

as the satellites observed in regular 4 circle diffraction experiment [1] were more or less of the same magnitude.

Let us note that the symmetry analysis showed that the iron atomic positions can be split into two orbits (four sites each), but because of the presence of two incommensurate modulations, the subdivision of the iron unit cell into two magnetic subsystems may be not unique.

Moreover, according to the most extreme theoretical magnetic models the iron magnetic moments can lie either in the basal plane or perpendicular to it. This situation, according to the both scenarios, was considered in the analysis of the results of previous neutron measurements [1]. We note that the extinction rules for the arrangement of iron magnetic moments slightly canted in the basal plane, as followed from our earlier observations, do not permit the appearance of magnetic satellites $(h \pm \varepsilon,\ k \pm \varepsilon,\ l)$ around l odd. Let us also note that the accumulated single crystal data are also not sufficient to exclude presence of a ferromagnetic component. This has to contribute e.g. to very strong (hkl) — all even peaks (200) and (220).

4. Conclusions

In light of recent results as well as the single crystal measurements [1] it seems to be certain that the magnetic structure cannot be treated as DCS system i.e. "flat" modulated one. It is likely that the magnetic symmetry may be extremely complicated double cone cycloid (DCC) one. As it was mentioned, we found no indication of the second phase transition as well as $(\varepsilon,\ \varepsilon,\ 0)$ and $(-\varepsilon,\ \varepsilon,\ 0)$ modulations (where $\varepsilon = 0.13$) exist. All this strongly calls for re-measuring both, the non-magnetic reflections that should clear up the situation connected with possible 2D- or 3D-cell-doubling and presence of the (0.13, 0.13, 0) magnetic satellites, which for unclear reasons were not observed in the experiment on VIVALDI.

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